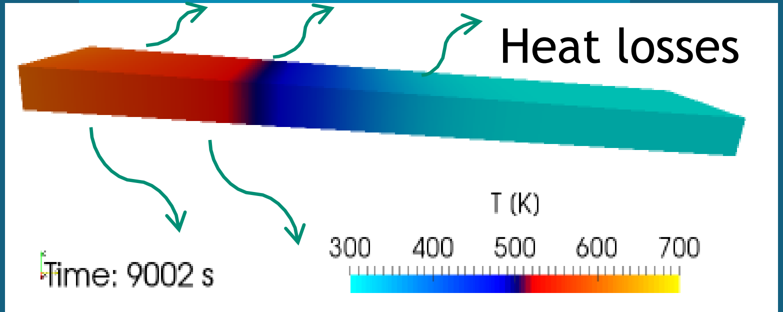
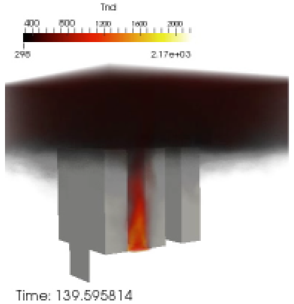
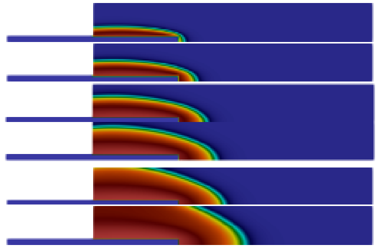
Sandia National Laboratories

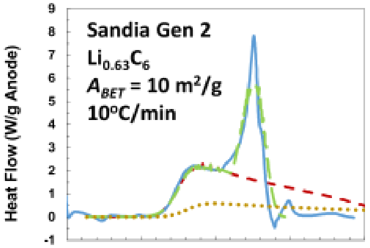
SAND2020-13738C

Predicting cell-to-cell failure propagation and limits of propagation in lithium-ion cell stacks









Presented by

Andrew Kurzawski, Loraine Torres-Castro, Randy Shurtz, Joshua Lamb, and John Hewson

U.S. DEPARTMENT OF ENERGY
NNSA

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Thermal runaway and cascading failure

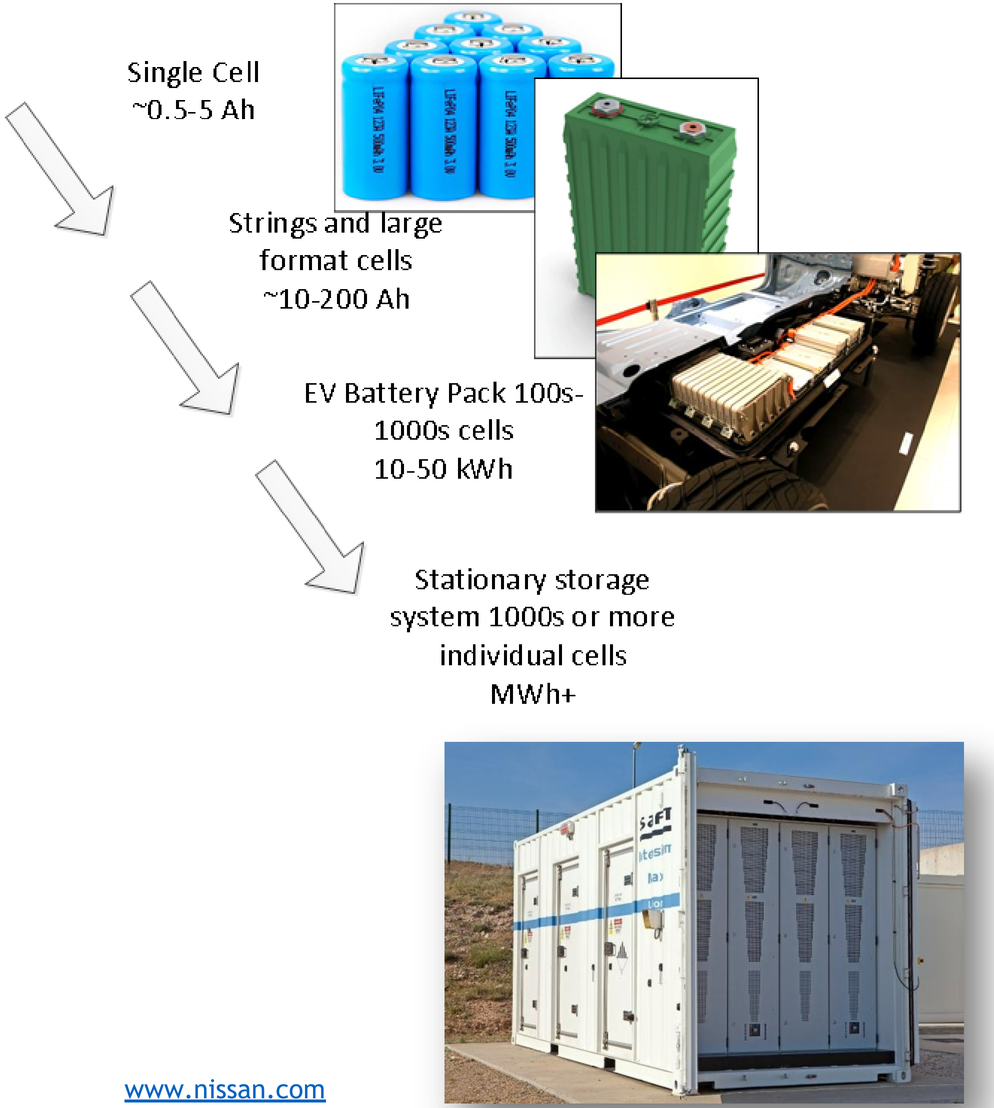
Validated reliability and safety is one of four critical challenges identified in 2013 Grid Energy Storage Strategic Plan

- Failure rates as low as 1 in several million
- Potentially many cells used in energy storage
- Moderate likelihood of 'something' going wrong

Increased energy densities and other material advances lead to more reactive systems

A single cell failure that propagates through the pack can have an impact even with low individual failure rates.

How do we decrease the risk?



Single Cell
~0.5-5 Ah

Strings and large
format cells
~10-200 Ah

EV Battery Pack 100s-
1000s cells
10-50 kWh

Stationary storage
system 1000s or more
individual cells
MWh+

www.nissan.com
www.internationalbattery.com
www.samsung.com
www.saft.com

Thermal runaway and cascading failure

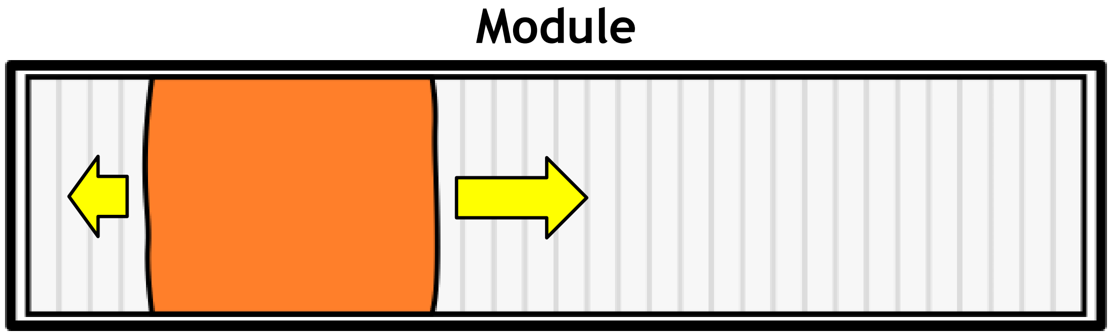
The current approach is to test our way into safety

- Large system (>1MWh) testing is difficult and costly.

Supplement testing with predictions of challenging scenarios and optimization of mitigation.

A key to designing safe systems at larger scales is understanding cascading thermal runaway.

Parallels can be drawn to thermal runaway of metal/oxidizer in cells to the spread of a premixed flame.



Source: (top) https://cmte.ieee.org/pes-essb/wp-content/uploads/sites/43/2019/06/2019-SM-UL-9540A-IEC-Lithium-Test-Summary.pdf
(bottom) https://www.ul.com/news/ul-9540a-battery-energy-storage-system-ess-test-method

Cascading failure testing with passive mitigation

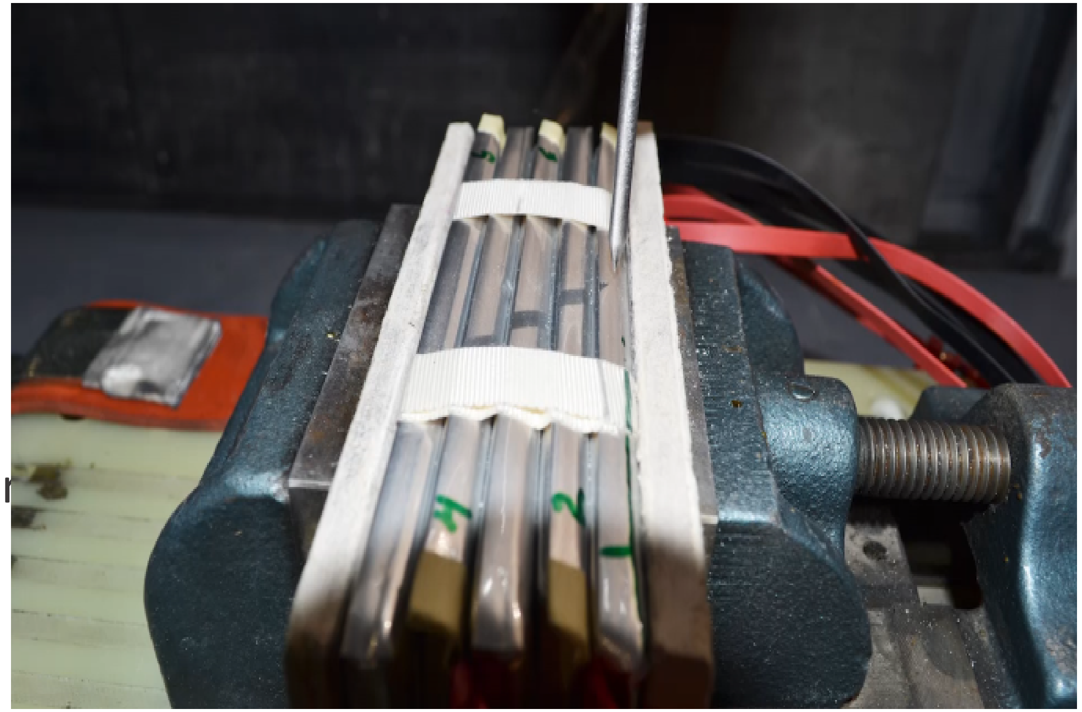
LiCoO$_2$ 3Ah pouch cells

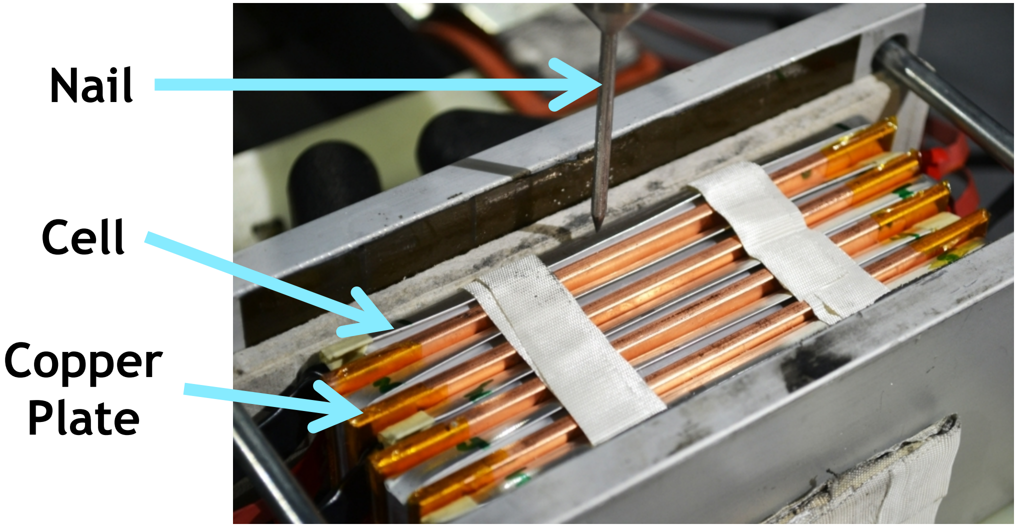
5 closely packed cells with/without aluminum or copper spacer plates

- Spacer thicknesses between 0.8 mm and 3.2 mm
- State of charge (SOC) between 50% and 100%

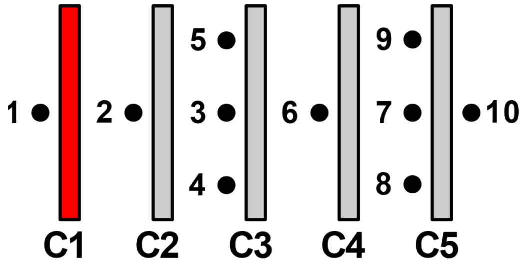
Failure initiated by a mechanical nail penetration in the outer cell (cell 1)

Thermocouples (TC) between cells and spacers (if present)

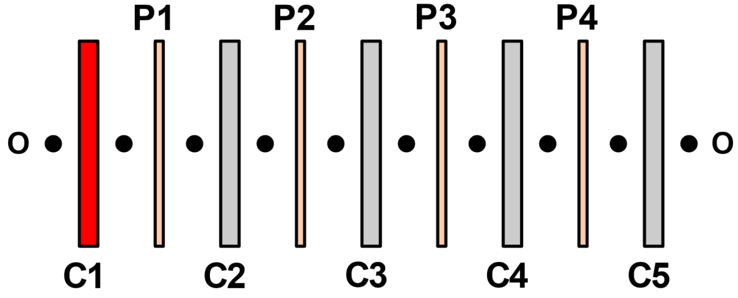




Thermocouple Locations without spacers



Thermocouple Locations with spacers



Finite element model for Li-ion cells in thermal runaway

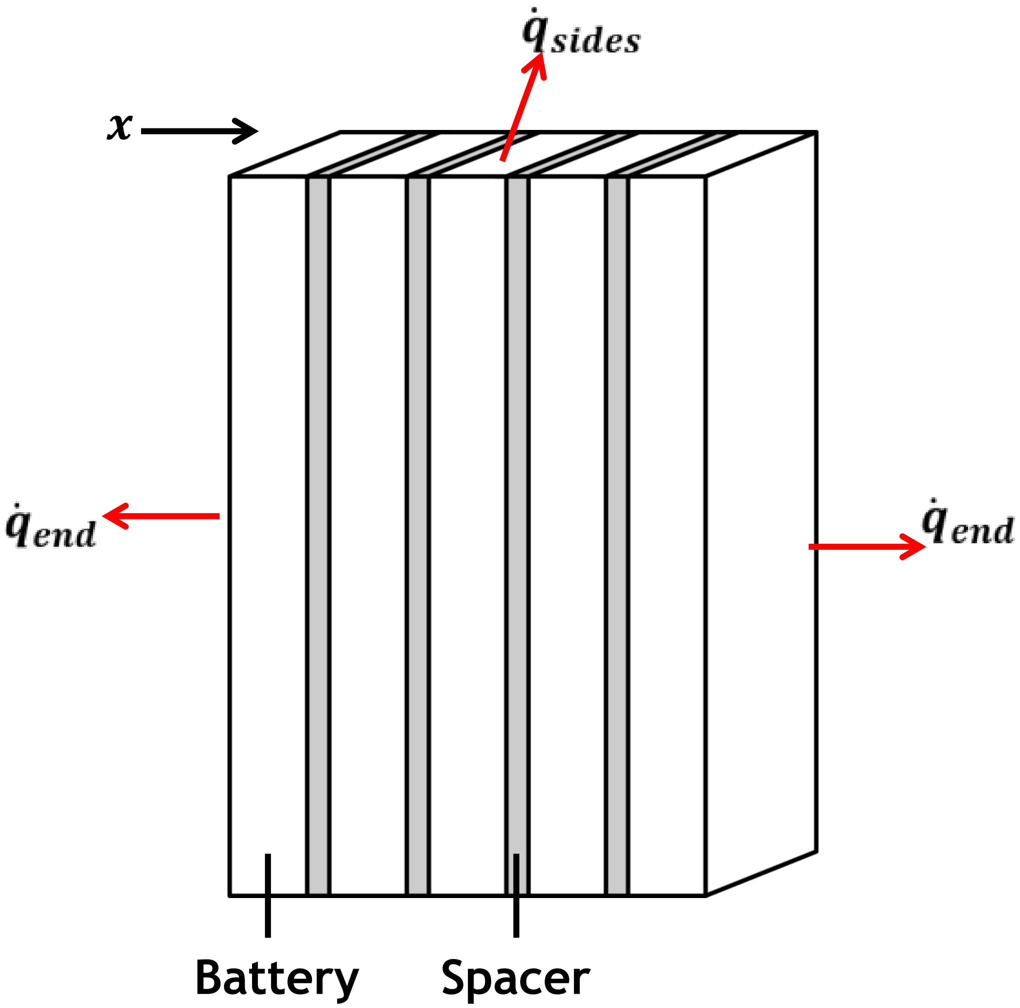
Discretization in one direction (***x***)

Modeled as a quasi 1-D domain of thin hexahedron elements

Multi-layered system
- Lumped battery material
- Spacers
- End block insulators

Convective heat transfer to surroundings (scaled by surface area to volume ratio for thin domain)

Heat conduction with chemical sources inside battery material



Finite element model equations

Energy conservation:

$$\rho c_p \frac{\partial T}{\partial t} = \nabla \cdot (K \nabla T) + \dot{q}'''$$

Mass conservation for species i with N_r reactions:

$$\frac{\partial \rho_i}{\partial t} = \sum_{j=1}^{N_r} (\nu_{ij}'' - \nu_{ij}') r_j$$

Energy source:

$$\dot{q}''' = -\sum_{j=1}^{N_r} \Delta H_j r_j$$

Chemical source terms for thermal runaway

Li-ion batteries contain a metal and oxidizer that can react with each other or alkyl carbonate electrolyte to release energy

These reactions occur at sub-grid scales and can be approximated as pre-mixed

Empirical chemical reactions:

- Short-circuit

$$C_6Li + CoO_2 \rightarrow C_6 + LiCoO_2$$

Metal and Oxidizer

- SEI decomposition (Richard 1999)

$$(CH_2OCO_2Li)_2 \rightarrow Li_2CO_3 + C_2H_4 + CO_2 + \frac{1}{2}O_2$$

- Anode-electrolyte (Shurtz 2018)

$$2C_6Li + C_3H_4O_3 \rightarrow 2C_6 + Li_2CO_3 + C_2H_4$$

Metal and Electrolyte

- Cathode-electrolyte (Hatchard 2001, Shurtz 2020)

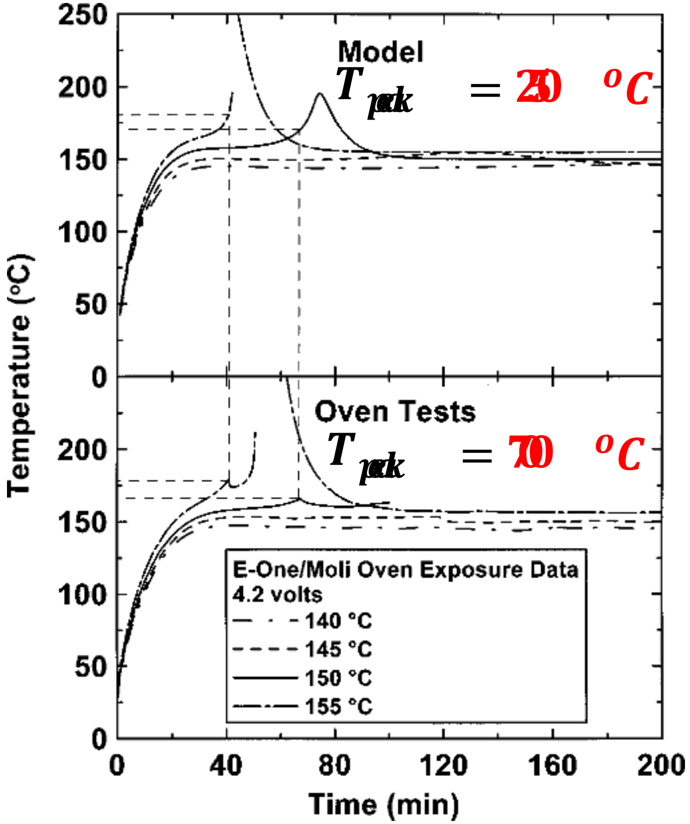
$$CoO_2 + \frac{2}{15}C_3H_4O_3 \rightarrow \frac{1}{3}Co_3O_4 + \frac{2}{5}CO_2 + \frac{4}{15}H_2O$$

Oxidizer and Electrolyte

Chemical source terms for thermal runaway

Preliminary chemistry models from literature

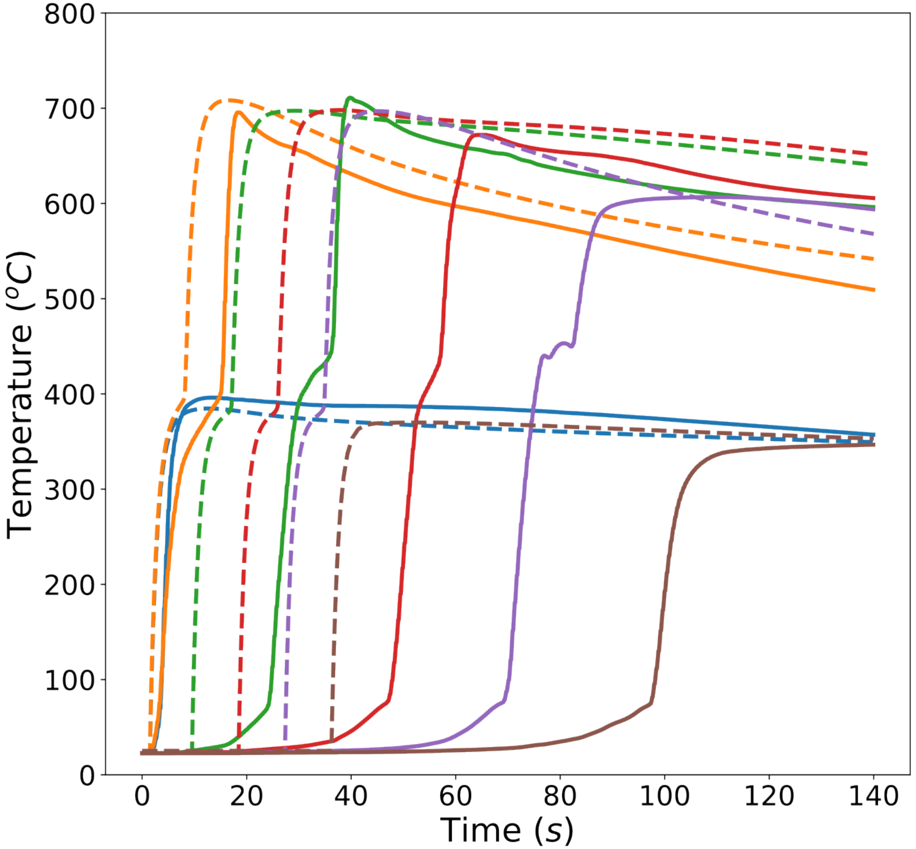
- Based on Dahn group (1999-2001)
- Calibrated for onset, but under-predicts peak temperature due to incomplete thermodynamics



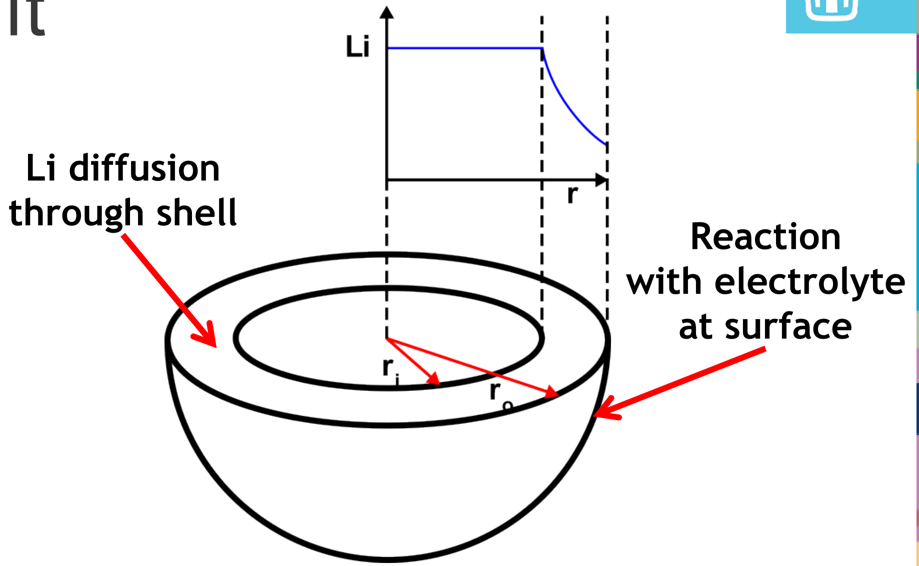
Hatchard, T. D., D. D. MacNeil, A. Basu and J. R. Dahn (2001). Journal of the Electrochemical Society **148**(7): A755-A761.

Extrapolating literature models to cell-scale with updated thermodynamics

- Reaction rates at propagation temperature (~700°C) are over-predicted
- Velocity of a premixed flame: $v \approx \sqrt{\dot{\omega}\alpha}$



Model for solid-state particle diffusion limit



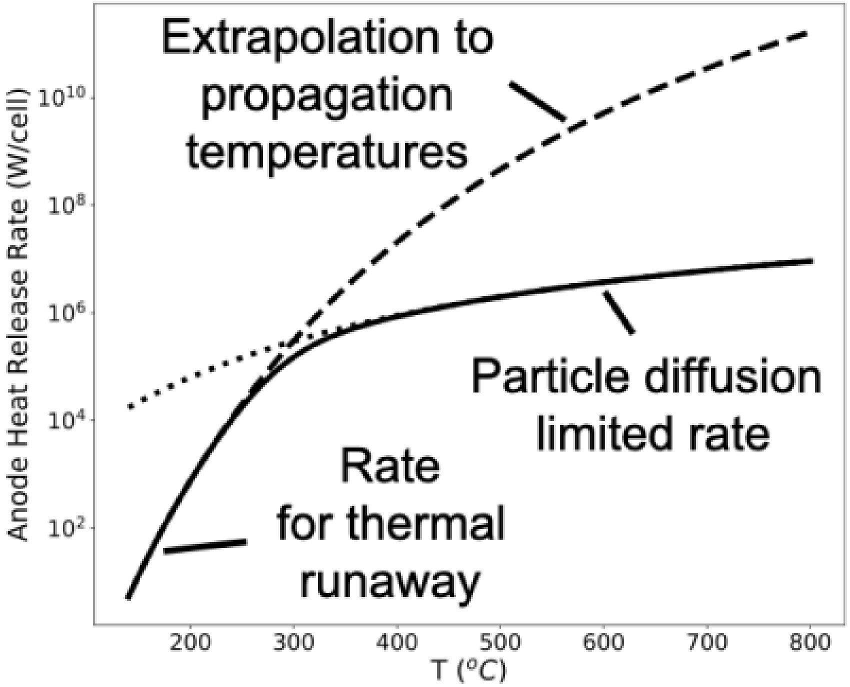
Challenge: Calorimetry measurements only at lower temperatures

- Lithium and oxygen must diffuse to the particle surface to react with the electrolyte.
- Serial reactions are corrected with the "Damköhler limited" form.

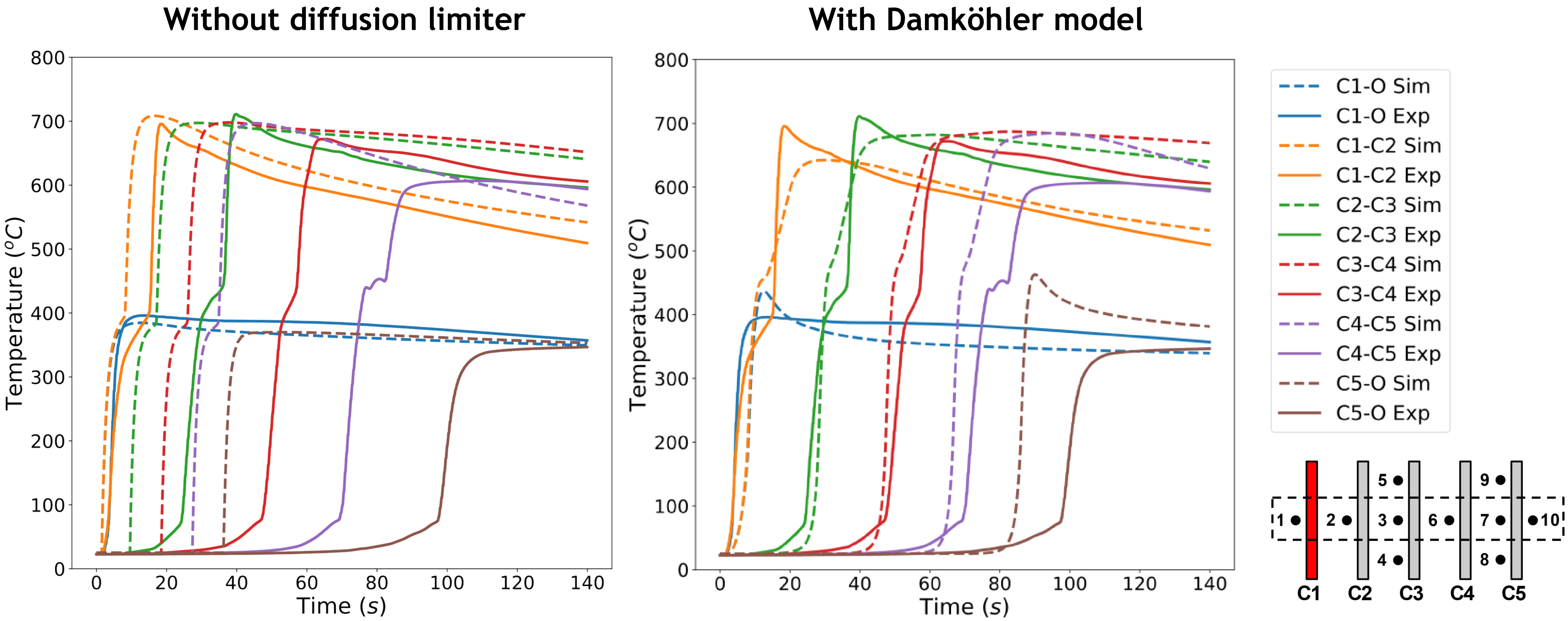
$$k' = \frac{k}{1 + Da}$$

- The Damköhler number is ratio of surface reaction rate to the rate of diffusion between an inner radius (r_i) and outer radius (r_o).

$$Da = \frac{A\exp\left(-\frac{E}{RT}\right)}{a_e \rho D_o \exp\left(-\frac{E_D}{RT}\right)} \frac{(r_o - r_i) r_o}{r_i}$$



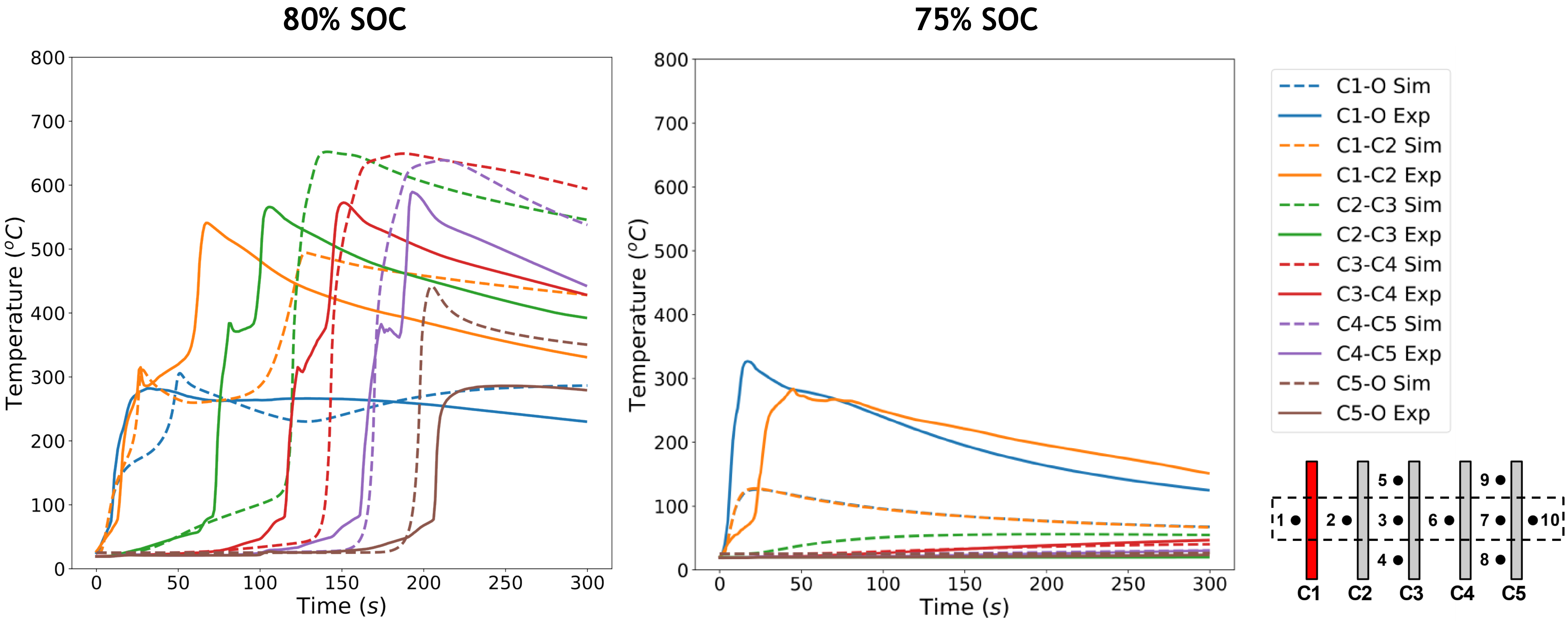
Five cell stack results: 100% SOC, no spacers



Without diffusion limiter

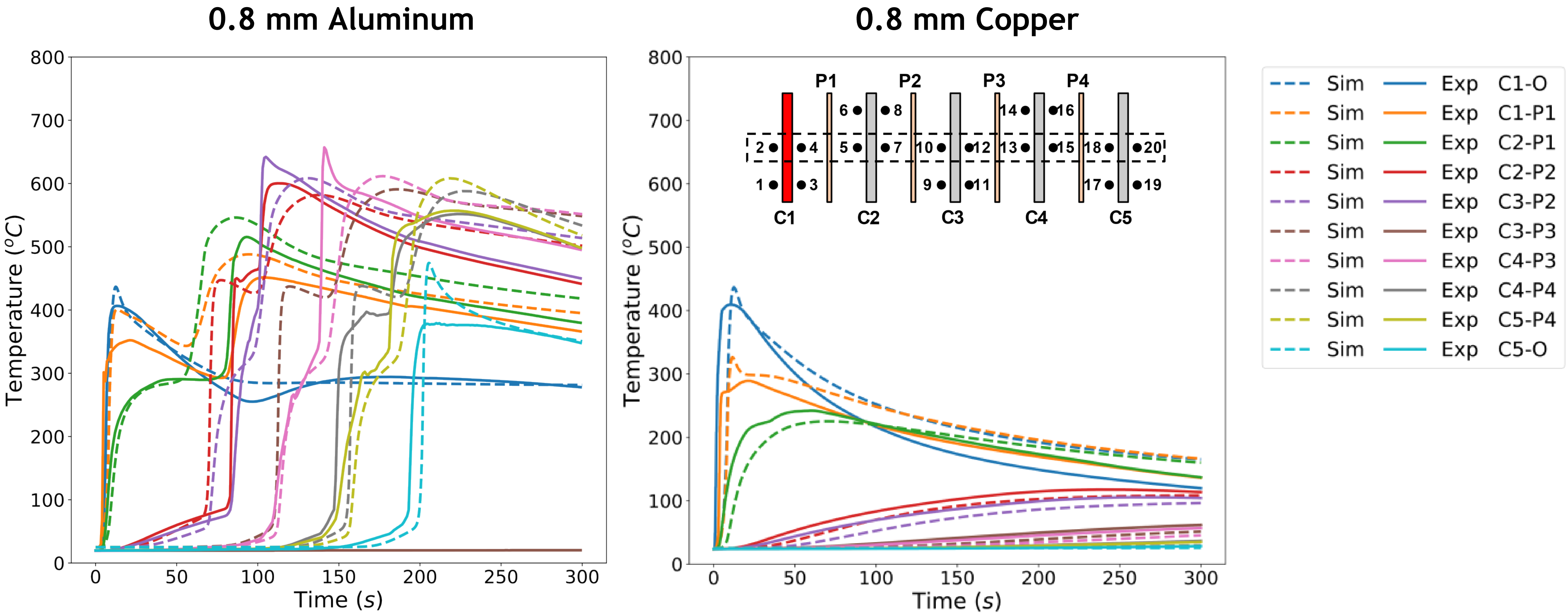
With Damköhler model

Results: lower state of charge (SOC)


80% SOC

75% SOC

Results: metallic spacers


0.8 mm Aluminum

0.8 mm Copper

Results: mitigation boundaries

Interplay between **heat capacity** of system and **energy release:**

$$\text{Energy/Capacity} = Q_{cells}/(m_{cells}c_{p,cells} + m_{spacers}c_{p,spacers})$$

Case Description	Energy/Capacity (K)	Propagation
100% SOC	940	Yes
0.8 mm Aluminum	819	Yes
0.8 mm Copper	778	Yes
80% SOC	752	Yes
1.6 mm Aluminum	725	No (Cell 2 Failure)
75% SOC	705	No (Cell 2 Failure)
1.6 mm Copper	663	No (Cell 2 Failure)
3.2 mm Aluminum	590	No
3.2 mm Copper	512	No
50% SOC	470	No

Summary

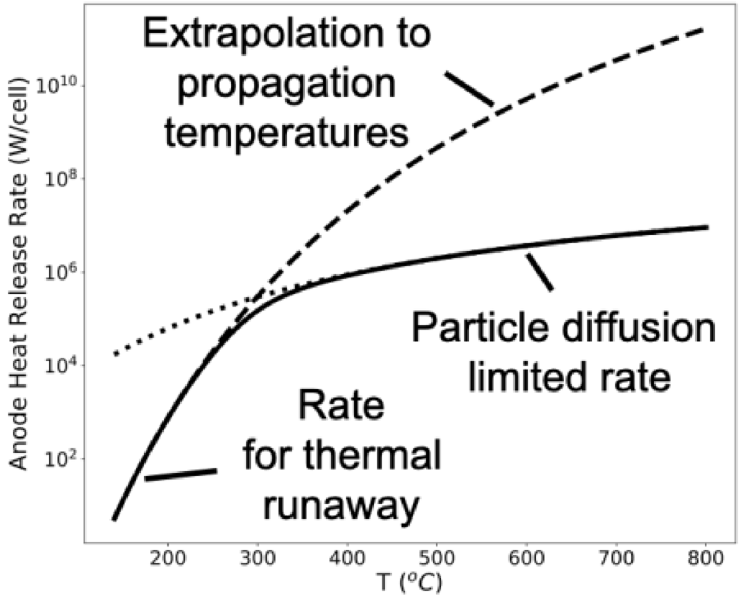
Using legacy chemistry source terms predicts onset of thermal runaway, but extrapolating this to higher temperature over-predicts cell-scale propagation speeds.

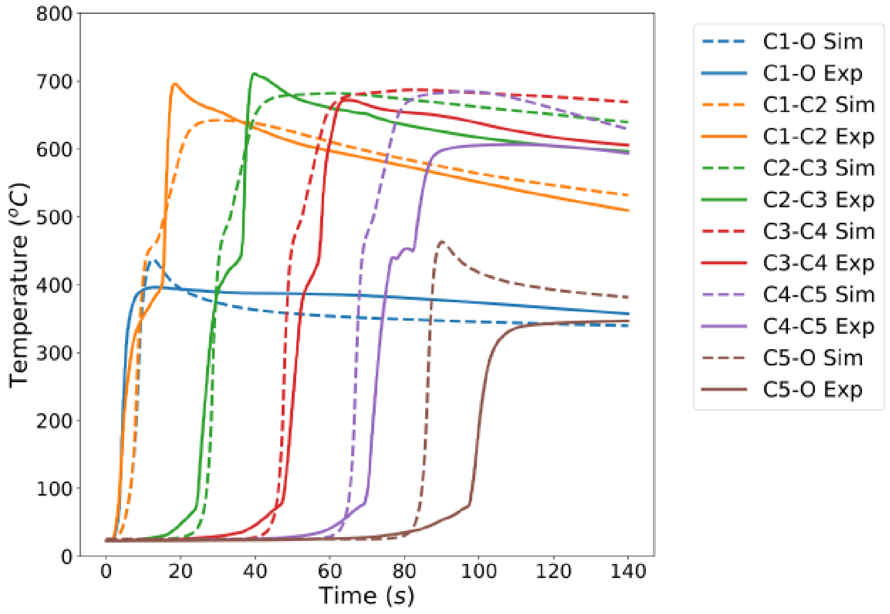
Results suggest that inclusion of intra-particle diffusion limits (or a similar change in the kinetics) becomes important for higher temperature cell-scale propagation.

Predictions were tested on a range of conditions with variable state-of-charge and passive mitigation spacers.

These results represent an extension of prediction capabilities to predict propagation and its limits over a range of thermal "dilution" conditions.

Understanding mitigation boundaries is important for designing safe energy storage systems.





Acknowledgements

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